We thank reviewer #2 for their comments, spotting several minor errors in the manuscript and the suggestions to improve the manuscript. We answer the reviewer's questions and comments below (our reply in blue, reviewer's comments in black).

Klein et al. measured the viscosity of particles with internally mixed NH_4NO_3 and sucrose using three techniques at atmospherically relevant humidity. The authors then predicted the viscosity based on mixing rules and the AIOMFAC-VISC model. They found that the mixing rule based on mole fractions is sufficient to predict the viscosity of a ternary system, e.g., the internally mixed NH_4NO_3, sucrose and H_2O. As viscosity is a very important property of aerosols and the observation data showing how the viscosity changes with particle composition (e.g, mixed inorganics and organics) are still very limited, this study is helpful in understanding the viscosity of internally mixed particles. However, I have some concerns about the authors' understanding of gas-particle partitioning. I recommend the publication of this study after the following comments could be addressed.

Major comments:

(1) My major comment arises from that the authors may have mixed the definition of mixing time and equilibrium time. Mixing time and equilibrium time are two different time scales. The equilibrium time refers to the time scale to achieve gas-particle equilibrium, affected by many factors, including particle viscosity, the volatility of partitioning compounds, particle size and concentrations etc, as gas-particle partitioning is controlled by gas-phase diffusion, interfacial transport, and particle-phase diffusion (Mai et al., 2015; Shiraiwa and Seinfeld, 2012; Li and Shiraiwa, 2019). However, the mixing timescale is mainly related to the particle-phase diffusion, as the authors wrote in Equation (2) in this study. Therefore, I suggest the authors be careful when using the term of "equilibrium time", e.g., the section of Appendix C, where the equilibration times are actually mixing timescales (Line 603, Line 616-619, Table C1).

We absolutely agree with the reviewer on the difference between equilibrium time and mixing time. We will carefully check and correct the revised manuscript in cases in which we had used equilibration time although we meant mixing time.

To clarify the wording in Appendix C: we add this sentence after line 605: "For these large particles mixing timescales should be equal to the equilibrium times scales due to the fast diffusion rate of water in the gas-phase and the high vapor pressure of water

(2) The authors applied three mixing rules, i.e., the mole-fraction based, the mass fraction based, and ZSR, to predict viscosity of the NH_4NO_3-sucrose-H_2O system. Many other studies also applied the Gordon-Taylor equation combined with the VTF equation to predict the viscosity of particles with mixed compounds (Dette and Koop, 2015; Li et al., 2020; O'brien et al., 2021; Koop et al., 2011). As the values of T_g of NH_4NO_3, sucrose, and H_2O are available, it is possible to estimate the viscosity based on the Gordon-Taylor equation. Would the predictions using the Gordon-Taylor equation agree with the predictions based on the mole-fraction based mixing rule?

This is an interesting suggestion. However, the problem with the VTF equation to estimate viscosity using Gordon-Taylor to estimate the glass transition of a mixture is that you need not just the glass transition temperatures for the pure compounds, but also need to estimate the Gordon-Taylor constant and for the VTF equation the fragility parameter. If you take those suggested by Li et al. (2020), namely a Gordon-Taylor constant of k_{GT}=2.5 and a fragility parameter of d=10, the prediction for an ammonium nitrate/sucrose mixture with OIR=4 overestimates the measured viscosity by about 3 orders of magnitude at an intermediate water activity of 0.5. It needs a lower fragility parameter of about 7 and a significantly larger Gordon-Taylor constant of about 6 to come close to the experimental data. See Fig. below:
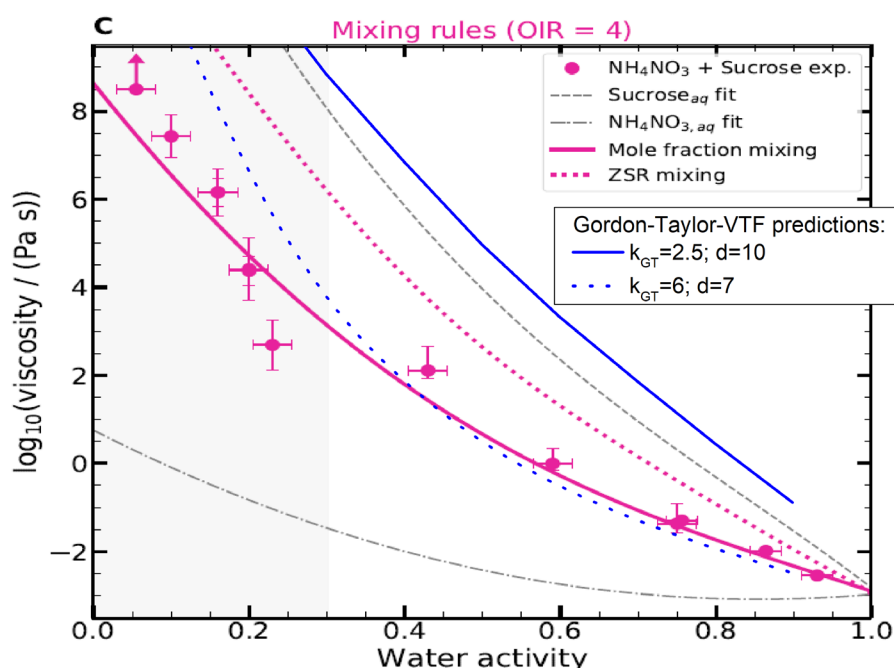
(3) In the section of 3.3 for Atmospheric Implications, the authors stated that "global models often assume equilibrium partitioning is achieved for fine particulate matter… if the mixing times exceed the chemical time step, it makes the quasi-instantaneous equilibrium assumption questionable". I do not agree with this statement. Note that current chemical transport models (CTMs) often assume SOA partitioning is rapid, i.e., instantaneous equilibrium partitioning (Pankow, 1994) is usually employed for the gas-particle partitioning of semi-volatile organic compounds (SVOCs) forming SOA particles. When particle is viscous, however, the equilibrium timescale of SVOCs can be longer than 1 h, in which case kinetic partitioning of SVOCs should be considered instead of instantaneous equilibrium partitioning (Li and Shiraiwa, 2019; Maclean et al., 2021; Zhang et al., 2024). For NH_4NO_3 partitioning, however, I think there is no argument questioning the assumption of instantaneous equilibrium of NH_4NO_3 partitioning currently applied in CTMs. Therefore, I am wondering the meaningfulness of calculating the mixing timescales of NH_4NO_3 partitioning in fine particles. In the abstract, the authors

should clearly state that they actually calculated the mixing time of NH$_4$NO$_3$ in internally mixed particles and in the paper title, the authors should clarify that equilibrium partitioning may be a reasonable assumption for NH$_4$NO$_3$ partitioning. In addition, at Line 314, the authors wrote "the mixing time scales with the square of the particle radius, regardless of the composition". This is not correct as the mixing timescales do relate to the composition because the bulk diffusion coefficients are different for different diffusing compounds.

We absolutely agree with the reviewer that when the particle is viscous the equilibrium time scale of SVOCs can be longer than 1 h and instantaneous equilibrium partitioning should no longer be used. That is what we intended to say when writing in line 306 ff: "Global models often assume equilibrium partitioning is achieved for fine particulate matter within the typical model time steps used for periodic output (tens of minutes to ca. 1 hour, e.g. Bian et al. (2017)). If the mixing times exceed the chemical time step in global models, it makes the quasi-instantaneous equilibrium assumption questionable." To make this clearer we will add "condensed phase" before "mixing times" in the last sentence.

We disagree with the reviewer that there is no argument questioning the assumption of instantaneous equilibrium of NH4NO3 partitioning. Imagine the following scenario: organic particles are in a polluted environment within the boundary layer and have sufficient time to reach equilibrium with ammonia and nitric acid at moderate temperatures. Now these particles experience a convective event with rapid updraft into the free troposphere where the gas phase concentrations of ammonia and nitric acid are low. If these organic particles containing ammonium nitrate would be sufficiently viscous (becoming more and more viscous with the lower temperatures of the updraft), the partitioning of ammonia and nitric acid back to the gas phase could potentially become kinetically limited. This is very similar to the scenario discussed in Bastelberger et al. (2017; Atmos. Chem. Phys., 17, 8453–8471), there for a semi-volatile organic instead of the equally semi-volatile ammonium nitrate. In the present manuscript we argue that this scenario indeed (as the reviewer writes) will not be likely to happen for ammonium nitrate as the viscosity of such mixed particles is quite small according to our measurements. But to our understanding this is not yet well established as measurements of the viscosity of inorganic/organic mixtures are scarce.

Referring to the question of the reviewer about the title: we indeed think that equilibrium partitioning is valid not only for ammonia and nitric acid but also for SVOCs as we show the addition of ammonium nitrate drastically reduces the viscosity of these particles as long as they are well mixed and do not exhibit phase separation.

We add the following two sentences at line 344 to make this clearer to the reader: "While the estimations for the mixing times are done for the ammonium ion, the same conclusion holds true for semi-volatile organic compounds (SOVCs) as long as the particles are single-phase and well mixed. They do not apply to the case of two-phase particles with an organic-rich phase."

We thank the reviewer for spotting that line 314 of the manuscript is misleading and apologize. We want to point out that for Fig. 7 we consider a single size only as mixing times scale with r^2 for a **fixed** composition. We will correct this incorrect phrasing in the revised manuscript.

Specific comments:

(1) Line 90: I could not find the SI file attached?

Thank you, we decided not to have a SI after all; in line 90 we should have referred to Appendix C. Corrected for the revised version.

(2) In the caption of Figure 3, is Tong et al. 2018 actually Tong et al. 2022?

Corrected for the revised version.

(3) Line 251-253, the authors concluded that the viscosity values reported in Tong et al. (2022) for NH_4NO_3- H_2O (Fig. 4) are questionable due to the volatilization of NH_4NO_3. Does this mean their data for the pure NH_4NO_3 showed in Fig. 3A are also questionable? Their data look comparable to Laliberté(2007) in Fig. 3A.

This is an interesting question. The reviewer is correct: the data of Tong et al. (2022) for the binary, aqueous ammonium nitrate agree with the La Liberté (2007) data. We cannot really assign what may have gone wrong with the Tong et al. (2022) experiment for the mixed system; see also the corresponding reply to the same question of reviewer 1. A simple explanation – which we are not able to prove – would be that the OIR was different from the reported one. We decided to delete the last sentence of this paragraph for the revised version.

(4) Line 258, in your system, what is the possible reason for the significant increase in viscosity below a_w of 0.1?

We do see a sudden increase of at least 5 orders of magnitude in viscosity only in the data for the OIR=1 system (cp. Fig. 4) below aw equal 0.1. As written in the manuscript we do not have any visual indication that NH_4NO_3 crystallized at this water activity, but we do not have any proof that this did not happen either. We will soften our statement for the revised manuscript by adding the following sentence to the paragraph: "However, we cannot prove that no crystallization of NH_4NO_3 occurred, causing the sudden increase in viscosity below aw equal 0.1 for the mixed droplets with OIR=1."

(5) Line 315-316, the authors calculated the diffusion coefficients of NH_4NO_3 via the Stokes-Einstein relation. However, previous studies have showed that the Stokes-Einstein relation is not suitable (with large underestimation) to predict the diffusion coefficients for such small diffusing molecules (Evoy et al., 2020; Price et al., 2015). Please also double check the calculation of D_{H2O} (Line 613) in Equation (C1).

As the reviewer writes, calculating diffusivity via Stokes-Einstein for small molecules potentially underestimates the diffusivity for small molecules, see e.g. in addition to the references mentioned by the reviewer also Bastelberger et al. (2017; Atmos. Chem. Phys., 17, 8453–8471). In the context of the atmospheric implications section such

underestimation will make mixing times even faster. This is stated on line 320 of the manuscript already, but we will add a sentence to the revised manuscript to strengthen the statement: "Therefore, the mixing times calculated using these derived diffusivities represent an upper limit." In addition, we will insert a new paragraph after the sentence.

(6) Line 200: "10^{12} Pa" should be "10^{12} Pa s".

Corrected.

(7) Caption of Figure 6: "the viscosities of NO_3–sucrose–H_2O" should be "$NaNO_3$–sucrose–H_2O".

Corrected.

(8) Table A1, what temperature the viscosity parameterization showed in Table A1 is suitable to?

Good point: this information is missing in the manuscript. The parameterizations in Table A1 refer to 293 K; added to manuscript.

(9) The mixing times showed in Fig. B2 are for which compound? The mixing times of different molecules are very different (refer to my major comment 3).

These are estimations for the mixing of the ammonium ion within aqueous toluene–derived SOA containing NH_4NO_3 for a dry mass ratio of organic to inorganic equal 2:1. We argue why we are using the ammonium ion radius in lines 315-325.

(10) Table C2, what d_{max} is? Is it the diameter of film?

That refers to the outer diameter of the droplet in the poke-flow experiments to estimate the time needed for equilibration with respect to RH. We will add a footnote to the table to clarify this meaning.